# Persistent organic pollutants (POPs) in mussels and oysters in the USA

Thomas P. O'Connor
*National Status and Trends Program*
*NOAA N/SCI1*
*1305 East West Hwy*
*Silver Spring MD 20910*
*USA*
*email tom.oconnor@noaa.gov*

## Abstract

**Since 1986 the NOAA National Status and Trends (NS&T) Program Mussel Watch has monitored concentrations of persistent organic pollutants (POPs) and other trace chemicals in the coastal United States by sampling mussels, oysters, and sediment. POPs have all been banned in the USA and concentrations in mollusks are decreasing. For some POPs there is a good correlation between concentrations in mollusks and numbers of people living near a collection site. Medians and 85$^{th}$ percentiles of USA data serve as a basis for comparison with similar data from programs in other nations.**

**Key words: Mussel Watch, Persistent organic pollutants, Monitoring**

## Introduction

To assess the effects of human activities on the quality of coastal and estuarine areas throughout the United States, the National Oceanic and Atmospheric Administration (NOAA) created the National Status and Trends (NS&T) Mussel Watch Project in 1986. The primary goals are to quantify the extent and geographic distribution of chemical contamination and to determine whether it is decreasing or increasing.

The need for this type of wide-scale monitoring was emphasized by U.S. National Research (NRC, 1990) which indicated that most of the marine environmental monitoring in the U. S. was compliance monitoring; i.e.. testing wastewaters and other materials prior to discharge or to performing measurements prescribed by regulation very near discharge points. Since compliance monitoring, by design, covers very small spatial scales, it is programs such as NS&T that are required to focus on wider public concerns.

## Chemicals Monitored

The elements and groups of organic compounds listed in Table 1 are measured by the NOAA NS&T Mussel Watch Project. All the groups of chlorinated organic compounds in Table 1 are listed in the recently completed international POPs treaty (Hogue, 2001). Hexachlorobenzene, mirex, and endrin are three other chemicals listed in the POPs treaty that are also measured by NS&T Mussel Watch. However, those three compounds are not discussed further here because in more than 67% of the samples they provided no analytic signal. Concentrations of individual organic compounds are summed into the groups shown in Table 1. When comparing results from separate programs it is important to note differences in what the groupings represent. For ∑PCB, 18 congener concentrations are added and then multiplied by two to conform with the correlation found between the sum of 18 and the sum of concentrations of PCB homologs (O'Connor, 1996).

Total DDT (∑DDT), total chlordane (∑Chlordane), and total dieldrin (∑Dieldrin) are chlorinated pesticides. All uses of DDT and dieldrin were banned in the United States in the 1970s. Chlordane use on U.S. crops ended in 1983, and its use for termite control effectively ended in 1988 (Shigenaka, 1990). Polychlorinated biphenyls (∑PCB) are chlorinated compounds first used in the 1920s for a number of industrial purposes. Their high heat capacities and low dielectric constants were ideal for use in electrical transformers and capacitors. PCB use in the United States began being phased out in 1971, and a ban on new uses took effect in 1976. Large changes in concentrations of ∑DDT and ∑PCB were seen at some locations in the 1970s following bans on their further use (Mearns et al., 1988). The compounds are still found in tissues and sediments because PCB-containing devices are still in use, chlordane remains in the ground as a termiticide, and ∑DDT remains in the environment due to its resistance to degradation (Woodwell et al., 1971). The pesticide DDT is metabolized to DDE and DDD in the environment, but those compounds degrade very slowly under environmental conditions.

**Table 1.**

Chemicals measured in mollusks

Elements
Ag, Al, As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Se, Zn

Concentrations of individual organic compounds have been aggregated into groups.

∑PCB= twice the sum of concentrations of eighteen congeners
PCB8, PCB18, PCB28, PCB44, PCB52, PCB66, PCB101, PCB105, PCB118, PCB128, PCB138, PCB153, PCB170, PCB180, PCB187, PCB195, PCB206, and PCB209

∑DDT= sum of concentrations of *ortho* and *para* forms of parent and metabolites
2,4'DDE; 4,4'DDE; 2,4'DDD; 4,4'DDD; 2,4'DDT; and 4,4'DDT

∑Chlordane= sum of concentrations of four compounds
*alpha*-chlordane, *trans*-nonachlor, heptachlor, heptachlorepobide

∑Dieldrin= sum of concentrations of two compounds
aldrin and dieldrin

∑BT= sum of concentrations of parent compound and metabolites
monobutyltin, dibutyltin, and tributyltin [concentrations in terms of tin]

∑LMW= sum of concentrations of twelve 2-and 3 ring PAH compounds
naphthalene, 2-methylnaphthalene, 1-methylnaphthalene, biphenyl, 2,6-dimethylnaphthalene, acenaphthene, acenaphthylene, 1,6,7-trimethylnaphthalene, fluorene, phenanthrene, 1-methylphenanthrene and anthracene

∑HMW= sum of concentrations of twelve 4-and more-ring PAH compounds, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*e*]pyrene, benzo[*a*]pyrene, perylene, dibenzanthracene, indeno[1,2,3-*cd*], pyrene, and benzo[*ghi*]perylene

∑PAH= LMW + HMW (sum of 24 PAH compound concentrations)

Polycyclic aromatic hydrocarbons (PAHs) are like elements in that they occur naturally. They are found in fossil fuels such as coal and oil and are produced when organic matter burns. Nonetheless, a multitude of human activities, from coal and wood burning to waste incineration, create PAH compounds in excess of those that would exist naturally. In addition, human production, transport, and use of oil release more PAHs to the environment, on a globally averaged basis, than does natural seepage (NRC, 1985). Because they are relatively more concentrated in oil than in combustion products, the low molecular-weight (LMW) 2- and 3-ring PAH compounds (Table 1) can be classified separately from the higher molecular-weight (HMW) 4- and 5-ring PAH compounds.

In recent years, the Mussel Watch project has added currently used pesticides and chlorinated hydrocarbons to its list of regularly measured monitored chemicals. National distributions of those compounds have been reported by Wade et al. (1998).

## Sampling Sites

The NS&T Mussel Watch Project is national in scale and sampling sites (Figure 1) should be representative of large areas rather than the small-scale patches of contamination commonly referred to as "hot spots." To this end, no sites were knowingly selected near waste discharge points. Furthermore, since the Mussel Watch Project is based on analyzing indigenous mussels and oysters, a site must support a sufficient population of these mollusks to provide annual samples.

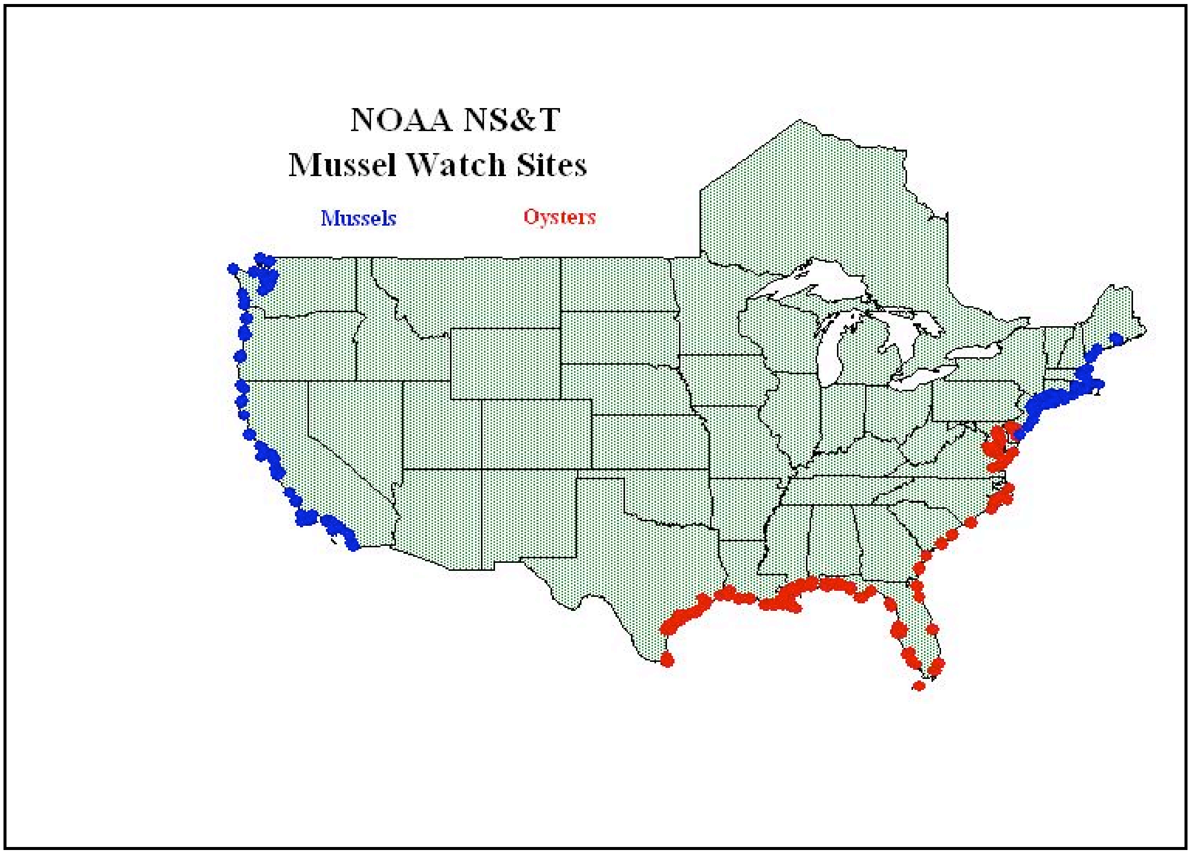


**Figure 1. NOAA NS&T Mussel Watch Sites**

NS&T sampling sites are not uniformly distributed along the coast. Within estuaries and embayments, they average about 20 kilometers (km) apart, while along open coastlines the average separation is 70 km. Almost half of the sites were selected in waters near urban areas, within 20 km of population centers in excess of 100,000 people. This choice was based on the assumptions that chemical contamination is higher, more likely to cause biological effects, and more spatially variable in these waters than in rural areas. There have been 238 marine site sampled over the period 1990 to1989. Lauenstein et al. (1997) provide a full description of each site.

## Species Collected

Since no single species of mollusk is common to all coasts, it has been necessary to collect seven different ones: the blue mussel *Mytilus edulis* on the East Coast from Maine to Cape May, NJ; the American oyster *Crassostrea virginica* from Delaware Bay southward and throughout the Gulf of Mexico; the mussels *M. edulis* and *M. californianus* on the West Coast; the oyster *Ostrea sandvicensis* in Hawaii; the smooth-edged jewel box *Chama sinuosa at* the one site in the Florida Keys; and the mangrove oyster *Crassostrea rhizophorae* in Puerto Rico

The multitude of species complicates comparisons among sites, because tissues of different species can have different chemical concentrations even if the surrounding environments are identical. At sites in Long Island Sound where it was possible to sample both mussels and oysters, *M. edulis* and *C. virginica*, three trace elements, Ag, Cu and Zn, were enriched in oysters by more than a factor of 10 relative to mussels. For other elements and for organic compounds no strong species-effect was observed. Similarly, at a site off the Columbia River two species of mussels, *M. edulis* and *M. californianus*, were sampled. In that case there were no important concentration differences for any element or any organic compound (O'Connor, 1994)

## Methods

Mollusks are shucked and whole soft parts are analyzed for their chemical content. Mollusks are collected at marine sites in the months of November through March with each site visited within 30 days of a prescribed date. In the Great Lakes, collections are made in August. From 1986 through 1991, six separate composite samples of 20 oysters or 30 mussels were collected at each site. Three composites were homogenized for trace element analyses and three for trace organic analyses. That protocol still applies whenever a site is sampled for the first time. Otherwise, since 1992 only two composite samples have been collected; one for organic and one for trace element analysis. The annual concentrations used in this report are the arithmetic means of three concentrations measured prior to 1992 or the single value measured since then. All data are available on the Internet at http://ccmaserver.nos.noaa.gov. The detailed methods for sampling and chemical analysis are contained in National Status and Trends Methods Documents (Lauenstein and Cantillo, 1993 a-d, and 1998).

## Trends

A temporal trend is a statistically significant correlation between contaminant concentration and year. Trends for 1986 through 1996 were reported by O'Connor (1998). For all the POPs groups listed in Table 1 annually measured concentrations are decreasing ($p<0.05$) at a great many individual sites while increasing at few or none and that annual national median concentrations are decreasing in the coastal United States.

## Summary statistics on concentrations

While they do not enter into trend analysis, actual concentrations are of interest. For all chemicals, the national distributions of concentrations are approximately lognormal. Central tendencies and variations are tabulated in Table 2 as the $15^{th}$, $50^{th}$, and 85th percentiles among 214 site means sites measured in 1990 at marine sites (O'Connor, 2001). That year was chosen as the base for comparison because it was the year in which the most sites were sampled.

**Table 2.**

$15^{th}$, $50^{th}$, and $85^{th}$ percentiles concentrations among 214 sites measured in 1990. PAHs are not POPs but are included in parts of the text

| Chemical | $15^{th}$pctl. | $50^{th}$ pctl. | $85^{th}$ pctl |
|---|---|---|---|
| ∑Chlordane | 6.2 ng $g^{-1}$(dry) | 13 ng $g^{-1}$(dry) | 34 ng $g^{-1}$(dry) |

| | | | |
|---|---|---|---|
| ∑Dieldrin | nd | 3.5 | 8.5 |
| ∑DDT | 12 | 31 | 140 |
| ∑PCB | 25 | 110 | 420 |
| | | | |
| ∑PAH | 77 | 230 | 1100 |
| ∑LMW | 48 | 110 | 320 |
| ∑HMW | 24 | 120 | 770 |

### Concentrations above public health advisories

The intent of the Mussel Watch Project is to monitor the status and trends of coastal contamination, regardless of whether chemical concentrations present a hazard to marine biota or to human consumers of seafood. Naturally, though, there is interest in hazard potential. The U. S. Food and Drug Administration (FDA) prohibits the interstate shipment and sale of seafood containing more than specified limiting concentrations for Hg and certain chlorinated hydrocarbons. Among the 4000 mussel and oyster samples analyzed in the Mussel Watch Program, the mean and median wet weight percentages have both been 88%. Multiplying by 7.5 to convert wet-weight based limits to a dry-weight basis (assuming 85% wet weight) indicates that no mollusks collected in any year exceeded the FDA limit for Hg or any POP except for ∑PCB at the one site in Buzzards Bay,

### Concentrations affecting mollusks

The Mussel Watch Project has not included any biochemical measures of response to chemical contamination. Histological slides were made of gonadal tissue to quantify the reproductive stage of the organisms and neoplasia and parasitic infestations were noted. Kim et al. (1998) could not connect any of the parasite observations with tissue concentrations of chemicals. Hillman et al. (1992) did note that the ∑PAH body burdens at the 18 sites where neoplasia was found in *M. edulis* were among the higher body burdens, but ∑PAH body burdens were higher at sites without neoplasia. Moreover, in a detailed correlative and experimental examination Krishnakumar et al. (1999) could not establish that hemic neoplasia, endemic to mussels in Puget Sound, is induced or promoted by PAHs or PCBs.

Field collected mollusks obviously do not harbor lethal residues of chemicals but it is noted that the higher of the observed molluscan concentrations of organic chemicals including POPs are of the order of 1 nM/g-wet which is 1000x lower than µM/g critical body residues of nonpolar organics associated with death of marine organisms (Kane Driscoll, et al. 1998).

Widdows et al (1995) measured hydrocarbon concentrations in and Scope for Growth of *M. edulis* held in cages along hydrocarbon gradients in waters of the United Kingdom. When all their data are viewed as a whole, effects began to appear at body burdens of about 100 ng/g-wet of 2 and 3 ring petroleum hydrocarbons. Their chemical methods did not discriminate among different LMW compounds and comparisons with Mussel watch concentrations are only approximations but 100 ng/g-wet corresponds to about 750 ng/-dry ∑ LMW. This is well above the Mussel Watch "high" concentration (Table 2) but has been exceeded at 31 sites in at least one year since 1990.

Krishnakumar et al. (1994) associated alterations in lysosomes of digestive cells in mussels with body burdens of PAHs and PCBs. In their case PAH compounds were measured just as they are in the NS&T Program. (Their∑LMW and ∑HMW totals included 5 and 1, respectively, fewer compounds than the totals used in this paper but overall differences are slight). At the four sites of their nine where no lysosomal alterations were found, ∑LMW and ∑HMW concentrations in *M. edulis* were 80 and 90 ng/g -dry or less. Mollusks at about half of the NS&T sites have ∑LMW and ∑HMW concentrations below those levels. The five sites where alterations were found included one with ∑LMW and ∑HMW concentrations above the highest ever measured at a Mussel Watch site. However at the fourth most contaminated of the five affected sites, the ∑LMW and ∑HMW concentrations were 820 and 320 ng/g, respectively. Concentrations close those defining "high" in Table 2. The lowest concentrations among the five affected sites were 100 (∑LMW) and 120 (∑HMW), close to the median Mussel Watch concentrations and little different from the concentrations in unaffected mussels. The ∑PCB tissue concentrations, like those of the PAHs, were near or above the Mussel Watch "high" at the four most contaminated sites. Also like PAHs, ∑PCB concentrations at one affected and four unaffected sites were near or below the Mussel Watch median. Among other chemicals measured by Krishnakumar et al. (1994) concentrations were generally less than the Mussel Watch "highs" and not consistently elevated among affected mussels relative to normal mussels.

The relative importance of PAHs is exemplified in the data of Gardinali and Wade (1998). At 85 NS&T Mussel Watch sites, mollusk tissues have been analyzed for dioxins (including 2,3,7,8 TCDD) and furans (both groups included in the POPs treaty) and three low-concentration planar PCBs (congeners 77, 126 and 166) not routinely measured by NS&T Mussel Watch. All these compounds and several PAHs can be compared on a common basis in terms of their ability to induce aryl-hydrocarbon hydroxylase activity in rat liver hepatocytes. Although the dioxins, furans, and planar PCBs are more potent inducers on a mole basis that the PAH compounds, the >1000 fold higher concentrations of PAHs causes that group to account for about 90% of the total induction equivalents in mollusk tissue.

## Human influence on concentrations

Concentrations of chemicals are evidence of contamination only if they would be lower in the absence of any human influence. There are fairly strong connections between human population density and chemical concentrations in oysters and mussels for ∑Chlordane, ∑DDT, ∑PCB, ∑butyltin, ∑HMW, and lead where Spearman correlation coefficients are greater than 0.5 (Table 3). The connection with urban areas for ∑PCB, and lead was also noted in the earlier Mussel Watch project (Goldberg et al., 1983) is not surprising. ∑Chlordane, ∑DDT, ∑PCB, and ∑butyltin, are synthetic chemicals whose concentrations would be zero in the absence of man. While ∑HMW and lead would always be found in mollusks, their present concentrations are due almost entirely to human actions. For ∑Dieldrin, ∑LMW and the elements Ag, Hg, and Zn the national-scale correlations (Table 3) are low but more than 40% of the “high” concentrations are found among the 15% of marine sites with 800,000 or more people living within 20 km.

**Table 3.**

Spearman correlation coefficients between molluscan concentrations and population for 1990 (n=number of sites=214). Population is the number of people living within 20 km of each site as per 1990 U.S. Census. (With so large an “n”, any coefficient with an absolute value greater than 0.135 is “significant” at the 0.05 level).

| Chemical | Spearman correlation coefficient ($r_s$) | | |
|---|---|---|---|
| ∑PCB | 0.623 | Cu(oyster) | 0.193 |
| Pb | 0.598 | Cr* | 0.181 |
| ∑BT | 0.585 | Hg | 0.179 |
| ∑Chlordane | 0.598 | Zn(mussel) | 0.174 |
| ∑DDT | 0.553 | ∑Dieldrin | 0.153 |
| ∑HMW | 0.520 | Ag (oyster) | 0.044 |
| Zn(oyster) | 0.486 | As | -0.024 |
| Ag (mussel) | 0.458 | Ni | -0.107 |
| ∑PAH | 0.473 | Se | -0.140 |
| Cu (mussel) | 0.288 | Cd | -0.312 |
| ∑LMW | 0.252 | | |

*Cr concentrations in correlation are site means for 1996-1998

## Comparisons among Mussel Watch Programs

In 1992 the United Nations Intergovernmental Oceanographic Commission (UN/IOC) inaugurated the International Mussel Watch Program by collecting and analyzing mollusks from 76 sites in Latin America. This was followed by a program in Asia in 1994-1997. The Latin American results are detailed by Farrington et al. (1994) and summarized by Sericano (1995). The Asia results have been collected by Tanabe (2000) in a volume containing the Tanabe et al. (1998) summary of the POPs data. In 1996 the United Nations Development Program (UNDP) and World Bank jointly sponsored a collection and analysis of samples in the Black Sea (Moore et al., 1998) Since 1979 the Reseau National d’Observation de la qualité du milieu marin (RNO) has been collecting and analyzing mussels and oysters from about 90 sites along the French Coast. The US and French results have been compared by Beliaeff et al. (1998) and in IFREMER (1996).

The median concentrations of POPs and lindane (a hexachloro- cyclohexane, HCH, pesticide not listed in the POPs Treaty) among all theses areas differ to some extent. Since HCHs are routinely used in India and in France for agricultural purposes the median concentrations are higher than elsewhere. PCBs are generally higher in the industrialized nations. The relative high median PCB for France in 1992 probably reflects the fact that PCB use was not banned until 1987. The high value for the Black Sea is the median of only five numbers.

**Table 4.**

Median concentrations of POPS in various Mussel Watch Programs

| Area (Program) | n | ∑DDT | ∑PCB | ∑Cdane | lindane |
|---|---|---|---|---|---|
| USA (NOAA/ NS&T) | 214 | 25 | 110 | 6.4 | 0.61 |
| France (RNO/IFREMER) | 90 | 12 | 96 [a] | | 3.3 |
| Latin America (UN/IOC) | 76 | 12 | 20 | 1.5 | |
| Black Sea (UNDP) | 6-13 | 80 | 200 | | |
| India (UN/IOC) | 19 | 47 [b] | 17 [b] | 0.21 [b] | 41 [bc] |
| Philippines (UN/IOC) | 13 | 10 [b] | 110 [b] | 20 [b] | 1.0 [bc] |
| Thailand(UN/IOC) | 21 | 18 [b] | 5.8 [b] | 4.2 [b] | 1.0 [bc] |

[a] IFREMER began measuring 7 PCB congeners in 1993. The median concentrations of the sum of the seven congeners over the years 1992,93, and 94 were 96 ng/g(dry) and 26 ng/g(dry) for France and the USA, respectively
[b] Numbers are 7.5x the numbers in the original citation which are in units of ng/g wet
[c] includes alpha and beta HCH as well as lindane (gammaHCH)

Sources
USA (NOAA/NS&T) all medians except lindane from O'Connor (2001), lindane from Beliaeff et a. (1998)
France (RNO/IFREMER) DDT and lindane medians from Beliaeff et al (1998), PCB median from RNO (1996)
Latin America (UN/IOC) medians from data in Farrington and Tripp (1995)
Black Sea (UN/IOC) medians from data in Moore et al. (1998)
India, Philippines, and Thailand (UN/IOC) medians from data in Tanabe et al (1998)

The upper ends of the concentration distributions differ more than the medians. The $85^{th}$ percentiles for NOAA/NS&T in Table 2 for ∑DDT, ∑PCB, and ∑Cdane are about twice as high as the corresponding $85^{th}$ percentiles in the Latin American data. Similarly, while the median concentration of the sum of seven PCB congeners was almost 4 times higher for the RNO (France) Mussel Watch than for the NS&T (U.S.), the $75^{th}$ percentiles were higher for the USA (RNO, 1996). Beliaeff et a. (2001) note that for 15 PAHs, the median concentrations for the U.S. and France were close to one another but at the $85^{th}$ percentile the U.S. concentrations were much higher. The fact the NOAA NS&T $85^{th}$ percentile concentrations tend to be high reflects the fact that about half the NS&T Mussel Watch sites are within 20 km of 100,000 or more people and this urban bias is evident at the high end of the national concentration distributions.

## Conclusions

While the main purpose of the NS&T Mussel Watch Project is to monitor temporal trends in coastal contamination, the data also define concentration distributions. Compared to results from other national programs, the median concentrations of POPs in mollusks in the US are slightly high and $85^{th}$ percentile much higher. The slightly high medians are probably due to the highly industrialized nature of the U. S. The higher upper end concentrations reflect the fact that there is a deliberate bias towards urban areas in the U.S. Mussel Watch site selection. Tissue concentrations in urban areas of two groups of organic chemicals, PAHs and PCBs, are in a range that may be causing a biological response the mollusks.

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